

ADHESIVE COMPOSITION

Patent Number: JP2000256633
Publication date: 2000-09-19
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Requested Patent: ☐ JP2000256633
Application Number: JP19990061516 19990309
Priority Number(s):
IPC Classification: C09J133/18; C09J11/04; C09J11/06; C09J133/14; C09J161/34; C09J163/00
EC Classification:
Equivalents:

Abstract

PROBLEM TO BE SOLVED: To provide adhesive compositions excellent in adhesion, heat resistance, and resistance to galvanic corrosion and, at the same time, having flame retardance in a non-halogen system.
SOLUTION: Adhesive compositions comprise (A) 70-90 pts.wt. copolymer of a polymerizable monomer having a nitrile group, a polymerizable monomer having an epoxy group, and another polymerizable monomer, (B) 7-20 pts.wt. epoxy resin, (C) 3-10 pts.wt. epoxy resin curing agent or curing accelerator and, based 100 pts.wt. components (A), (B), and (C) in total, (D) 20-40 pts.wt. nitrogen based compound, (E) 20-40 pts.wt. phosphorus based compound, and (F) 70-90 pt.wt. inorganic filler containing water of crystallization.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]

[The technical field to which invention belongs] Fire retardancy is acquired by the non-halogen system, and this invention relates to the suitable adhesives constituent for adhesives and an adhesion sheet excellent in an adhesive property, thermal resistance, and electric corrosion-proof nature.

[0002]

[Description of the Prior Art] In recent years, with the miniaturization of various electronic equipment, the loading density of electronic parts also becomes high and the property required of a patchboard or a semiconductor package used for it is also diversified. Thus, wiring occupancy area becomes small and, as for a printed wired board, the demand of the formation of a multilayer-interconnection board, the formation (FPC) of a pre KISHIBURU patchboard, etc. has been increasing increasingly. Although various adhesives or adhesive films were used for these patchboards in the manufacturing process, there was no adhesives or adhesion sheet which is excellent in an adhesive property, thermal resistance, and electric corrosion-proof nature, and has fire retardancy by the non-halogen system. Although what uses the dehydration coalification of the so-called "free radical trap effect" and oxy-halogenation antimony by carrying out combined use addition of an antimony system compound and the halogen system compound is known, problems, such as environmental pollution at the time of combustion, have not solved the old fire-resistant adhesives constituent.

[0003]

[Problem(s) to be Solved by the Invention] Invention claims 1 and 2 and given in three offers the adhesives constituent which is excellent in an adhesive property, thermal resistance, and electric corrosion-proof nature, and has fire retardancy by the non-halogen system.

[0004]

[Means for Solving the Problem] The copolymer 70 of the polymerization nature monomer in which this invention has the (A) nitrile group, the polymerization nature monomer which has an epoxy group, and other polymerization nature monomers - 90 weight sections, (B) An epoxy resin 7 - 20 weight sections, (C) epoxy resin curing agent or a hardening accelerator 3 - 10 weight sections, (A) It is related with the adhesives constituent which comes to blend the inorganic bulking agent 70 which contains (D) nitrogen system compound 20 - 40 weight sections, (E) Lynn system compound 20 - 40 weight sections, and (F) water of crystallization to the total amount 100 weight section of +(B)+(C) - 90 weight sections.

[0005] Moreover, the copolymer of the (A) component consists of 15 - 35 % of the weight of acrylonitrile, 0.5 - 4 % of the weight (meta) of metaglycidyl acrylate, and 61 - 84.5 % of the weight of other polymerization nature monomers, and this invention relates to the aforementioned adhesives constituent whose glass-transition-point temperature weight average molecular weight is 700,000 or more, and is -5 degrees C or less. Moreover, this invention relates to the adhesives constituent according to claim 1 which is a phenol resin constituent with which the nitrogen system compound of the (D) component consists of the mixture or the condensate of phenols, the compound which has a triazine ring, and aldehydes.

[0006]

[Embodiments of the Invention] As a polymerization nature monomer which has the nitrile group of the material which constitutes the (A) component in this invention, acrylonitrile etc. is mentioned, for example. As a polymerization nature monomer which has an epoxy group, metaglycidyl acrylate (meta) etc. is mentioned, for example. As other polymerization nature monomers, moreover, a methyl acrylate (meta), an ethyl acrylate (meta), A butyl acrylate, isobutyl acrylate (meta), acrylic-acid (meta)

2-ethylhexyl, (Meta) Acrylic-acid (meta) alkyl ester and acrylic-acid (meta) cycloalkyl ester, such as acrylic-acid lauryl and acrylic-acid (meta) cyclohexyl, (Meta) The acrylic acid (meta) containing a carboxyl group, the acrylamides (meta) containing an amide group, etc., such as metaglycidyl acrylate (meta) alkyl ester containing a glycidyl group and acrylic-acid (meta) hydroxyalkyl ester containing a hydroxyl group, are used. An acrylic acid (meta) means an acrylic acid and a methacrylic acid above. [0007] The copolymer of a component desirably 15 - 35 % of the weight of acrylonitrile (A) 20 - 30 % of the weight, Desirably 0.5 - 4 % of the weight of glycidyl (meta) acrylate 1.0 - 3.0 % of the weight, and 61 - 84.5 % of the weight of other polymerization nature monomers, It consists of 67 - 79 % of the weight desirably, and it is desirable from the point of an adhesive property, thermal resistance, flexibility, and intensity that weight average molecular weight is [800,000-1 million, and - 5 degrees C or less of glass-transition-point temperature] -10--30 degrees C desirably 700,000 or more.

[0008] The amount of the above-mentioned acrylonitrile used tends to become insufficient [15 % of the weight / thermal resistance], and if it exceeds 35 % of the weight, it will tend to become inadequate [adhesive strength]. At less than 0.5 % of the weight, the amount of the above-mentioned (meta) metaglycidyl acrylate used tends to become inadequate [solder thermal resistance], and if 4 weight sections are exceeded, it will tend to become inadequate [adhesive strength].

[0009] When there is an inclination for weight average molecular weight (value which measured by the gel permeation chromatography method (GPC), and was converted using the calibration curve by standard polystyrene) to become insufficient [less than 700,000 / flexibility or intensity] and molecular weight becomes not much high too much, there is an inclination for viscosity to become high and to become inadequate [workability]. The monomer concentration at the time of a solution polymerization start, the kind of polymerization initiator, and polymerization temperature can adjust molecular weight. Glass-transition-point temperature will tend to become inadequate [an adhesive property], if it exceeds -5 degrees C.

[0010] (A) The copolymer of a component is obtained by carrying out the solution polymerization of the polymerization nature monomer which has a nitrile group, the polymerization nature monomer which has an epoxy group, and other polymerization nature monomers by the well-known radical polymerization method. In this case, ester solvents, such as ketone system solvents, such as aromatic system solvents, such as toluene and a xylene, a methyl ethyl ketone, and a methyl isobutyl ketone, ethyl acetate, and butyl acetate, can be used as an organic solvent. Moreover, as a polymerization initiator, azobis system compounds, such as organic peroxide, such as benzoyl peroxide, dicumyl peroxide, dibutyl peroxide, t-butyl peroxide, and oxybenzoate, azobis isobutyl nitril, and azobisiso valeronitrile, can be used.

[0011] As for the epoxy resin of the (B) component in this invention, less than 5000 epoxy resin is preferably used for number average molecular weight by two or more organic functions. For example, liquefied epoxy resins, such as the bisphenol A type and a bisphenol female mold, Polyfunctional epoxy resins, such as a phenol novolak type and a cresol novolak type, can be used. Tradename D.E.R.330 of tradename Epicoat 807 of oil-ized shell epoxy incorporated company, Epicoat 827, Epicoat 828, and Dow Chemical Japan, Inc., D.E.R.331, D.E.R.361, the tradenames YD128 and YDF170 of **** Chemicals incorporated company, Tradename EPPN-201 of Nippon Kayaku Co., Ltd., EOCN-1012, EOCN-1025, tradename ESCN-001 of Sumitomo Chemical Co., Ltd., and ESCN-195 can be used.

[0012] Moreover, the (B) epoxy resin and a phenoxy resin with compatibility can be used, replacing them by some epoxy resins. As a phenoxy resin, tradename FENOTOTO -40 of **** Chemicals incorporated company, FENOTOTO YP-50, and FENOTOTO YP-60 can be used, for example.

[0013] Although not limited especially as the curing agent or hardening accelerator of the (C) component in this invention For example, anhydrous hexahydrophthalic acid and anhydrous methyl hexahydrophthalic acid, Acid anhydrides, such as an anhydrous methyl tetrahydrophthalic acid, bisphenol A, Tetrabromobisphenol A, a TORIMECHI roll allyloxy phenol, Phenols, such as a phenol novolak resin, a cresol novolak resin, and butyl-ized phenol resin, Aliphatic polyamine, aromatic polyamine, the 2nd class or tertiary amine, a melamine, a triazine compound, an organic acid, imidazole derivatives, a dicyandiamide, a triphenylphosphine, the poly mercaptan, etc. can be used. these are independent -- or more than one are mixed and optimum dose use can be carried out

[0014] (A) The loadings of a copolymer are 70 - 90 weight section. this -- if out of range, solder thermal resistance and a heat-resistant adhesive property fall

(B) The loadings of an epoxy resin are 7 - 20 weight section, and are 10 - 15 weight section

preferably. Solder thermal resistance falls under in 7 weight sections. If 20 weight sections are exceeded, an adhesive property will fall.

(C) The loadings of a curing agent and a hardening accelerator are 3 - 10 weight section. Solder thermal resistance and a heat-resistant adhesive property fall under in 3 weight sections. If 10 weight sections are exceeded, an adhesive property will fall.

(A) A component, the (B) component, and the (C) component are blended so that these total amounts may serve as the 100 weight sections.

[0015] As a nitrogen system compound of the (D) component in this invention Although it is not limited especially if it is the compound which has a triazine ring With the compound which has a triazine ring, for example, a melamine, benzoguanamine, The melamine resin which is the condensate of a SUPIRO guanamine, etc. and aldehydes, an acetaldehyde and formaldehyde, Some or all of an acetoguanamine resin, a benzoguanamine resin, spiro guanamine resins, etc. and those methylol machines A methanol, The thing and further which were etherified by lower alcohols, such as a butanol, a cyanuric acid, The phenol resin constituent which consists of the mixture or the condensate of cyanuric-acid derivatives, such as an ethyl SHIANU rate and an acetyl SHIANU rate, phenols and the compound which has a triazine ring, and aldehydes is mentioned, and thermal resistance and an adhesive point to the aforementioned phenol resin constituent is desirable. As the aforementioned phenols, polyhydric phenols, such as alkylphenols, such as a phenol, cresol, a xylene, ethylphenol, and butylphenol, bisphenol A, Bisphenol F, Bisphenol S, a resorcinol, and a catechol, etc. are mentioned, for example.

[0016] (D) The amount of the nitrogen system compound used is 25 - 35 weight section preferably in 20 - 40 weight section to the total amount 100 weight section of a (A)+(B)+ (C) component. Fire retardancy falls under in 20 weight sections. If 40 weight sections are exceeded, solder thermal resistance will fall.

[0017] An organic-phosphorus system compound can be used as a Lynn system compound of the (E) component in this invention. For example, the phosphoric ester of phosphoric ester, such as triphenyl phosphate, triglycidyl phosphate, a poly phosphate compound, a resorcinol poly phosphate compound, trixylenyl phosphate, and aromatic condensation phosphoric ester, and a biphenyl system is mentioned. (E) The amount of the Lynn system compound used is 20 - 40 weight section to the total amount 100 weight section of a (A)+(B)+ (C) component. Fire retardancy falls under in 20 weight sections. If 40 weight sections are exceeded, an adhesive property and solder thermal resistance will fall.

[0018] Moreover, phosphazene compounds, such as a compound which has the vinyl-group content alkyl ester which has a phosphazene ring as a compound which has both the nitrogen system compound which is the (D) component and the (E) component in this invention, and the Lynn system compound, and a phosphazene ring and the benzene ring, can be used.

[0019] An aluminum hydroxide and a magnesium hydroxide are mentioned as an inorganic bulking agent containing the water of crystallization of the (F) component in this invention. (F) The amount of the inorganic bulking agent used is 80 - 90 weight section preferably in the 70 - 100 weight section to the total amount 100 weight section of a (A)+(B)+ (C) component. Fire retardancy falls under in 70 weight sections. If the 100 weight sections are exceeded, an adhesive property will fall.

[0020] You may blend suitably a coupling agent and a pigment better known than before, a leveling agent, a defoaming agent, an ion trap agent, etc. if needed other than each above-mentioned component.

[0021]

[Example] An example explains this invention concretely below. In addition, the loadings in the following publications are expressed with the weight section.

[0022] The compound I shown in Table 1 and 2 is put into the reactor which equipped with the [manufacturing method of copolymer A-I] mixer, and the condensator, it heats at 80 degrees C - 85 degrees C, the compound II shown in Table 1 and 2 is added, it is kept warm for 4 to 8 hours, and the polymer made to react 20 to 40% by conversion is obtained. The methanol after cooling is added, polymer is settled and a supernatant is removed. The methanol which remained into polymer was dried, and the methyl ethyl ketone was added so that a solid content might become 15% continuously. The loadings (section) of Table 1 and 2 show the weight section.

[0023] The weight average molecular weight of this polymer A-I was measured on the conditions shown below by the gel-permeation-chromatography method (GPC), and was converted using the calibration curve by standard polystyrene.

<GPC conditions> -- use device: -- Hitachi 635 type HPLC [Hitachi Make]
 column: -- gel pack R440+R450+R400 -- M [the tradename by Hitachi Chemical Co., Ltd.]
 eluate: -- tetrahydrofuran measurement temperature: -- 40-degree-C flow rate: -- 2.0 ml/min detector: -
 - a differential-refractometer determination-of-molecular-weight result and glass-transition-point
 temperature are shown in Table 1 and 2 In addition, glass-transition-point temperature is calculated
 value. (Reference data; a guide for paints to synthetic resin, Kyoza Kitaoka work)

[0024]

[Table 1]

表 1

試 料 名		A	B	C	D	E	F	G
配合物 I (部)	メタクリル酸グリシジル	2	2	2	0.5	4	2	2
	アクリロニトリル	15	25	35	25	25	25	25
	n-ブチルアクリレート	30	53	53	54.5	51	28	67
	エチルアクリレート	53	20	-	20	20	45	-
	2-エチルヘキシルアクリレート	-	-	10	-	-	-	6
	メチルエチルケトン	5	5	5	5	5	5	5
配合物 II部	メチルエチルケトン	2	2	2	2	2	2	2
	t-ブチルパーオキシベンゾエート	0.05	0.05	0.05	0.05	0.05	0.05	0.05
重量平均分子量 ($\times 10^4$)		91	90	90	90	89	90	91
ガラス転移点温度 (°C)		-20	-20	-20	-20	-20	-10	-30

[0025]

[Table 2]

表 2

試 料 名		H	I	J	K	L	M
配合物 I (部)	メタクリル酸グリシジル	2	2	2	-	10	2
	アクリロニトリル	25	25	25	25	25	25
	n-ブチルアクリレート	53	53	53	55	65	-
	エチルアクリレート	20	20	20	20	-	73
	2-エチルヘキシルアクリレート	-	-	-	-	-	-
	メチルエチルケトン	10	-	5	5	5	6
配合物 II部	メチルエチルケトン	2	2	20	2	2	2
	t-ブチルパーオキシベンゾエート	0.05	0.05	0.1	0.05	0.05	0.05
重量平均分子量 ($\times 10^4$)		81	99	39	89	95	85
ガラス転移点温度 (°C)		-20	-20	-20	-20	-20	2

[0026] The material (*1-*10) shown in Table 3 and 4 to examples 1-9 and the copolymer obtained example of comparison 1-4 was added, and the varnish of an adhesives constituent was obtained. It applied so that the thickness after drying the obtained adhesives varnish on a polyimide film (35 micrometers in thickness) might be set to 70 micrometers, and it dried for 5 minutes at 140 degrees C, and the polyimide film with an adhesives layer was produced.

[0027] In addition, *1 - *10 material in Table 3 and 4 is shown below.

*1: Bisphenol A type epoxy resin (tradename by Epicoat 828:oil-ized shell epoxy incorporated company)

*2: Cresol novolak type epoxy resin (tradename by EPOTOTO YDCN703:**** Chemicals incorporated company)

*3: Phenol resin (tradename by ply OFEN LF2822:Dainippon Ink & Chemicals, Inc.)

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*4: Imidazole system hardening accelerator (tradename by cure ZORU 2 PZ-CN:Shikoku Chemicals incorporated company)

*5: Hydroxylation aluminum (tradename by HAIJI light H-42STE:Showa Denko K.K.)

[0028] *6: Biphenyl type phosphoric ester (tradename by CR-747:large 8 chemical-industry incorporated company)

*7: Methylation melamine resin (tradename by MERAN 523:Hitachi Chemical Co., Ltd.)

*8: Melamine phenolic resin (Hitachi Chemical Co., Ltd. make)

*9: Phenoxy resin (tradename by FENOTOTO YP-50:**** Chemicals industrial incorporated company)

*10: Acrylonitrile-butadiene rubber (tradename by PNR-1:JIEI S R incorporated company)

[0029] The polyimide film with an adhesives layer was produced like the example except having made the copolymer 533 weight section of example of comparison 5 example 1 change (combination 80 weight section) at the phenoxy resin (*9).

[0030] The polyimide film with an adhesives layer was produced like the example except having made the copolymer 533 weight section of example of comparison 6 example 1 change (combination 80 weight section) at acrylonitrile-butadiene rubber (*10).

[0031] The polyimide film with an adhesives layer was produced like the example except having removed hydroxylation aluminum, biphenyl type phosphoric ester, melamine resin, and the melamine phenolic resin from combination of example of comparison 7 example 2.

[0032] An adhesive property, solder thermal resistance, fire retardancy, a heat-resistant adhesive property, and electric corrosion-proof nature were investigated using the obtained polyimide film with an adhesives layer. The aforementioned property was investigated by the method shown below. The result is shown in Table 5 and 6.

[0033] <Adhesive property> The roll lamination was carried out heating a polyimide film with an adhesives layer at 120 degrees C on rolling copper foil (glossy surface) or a polyimide film, heat hardening was carried out at 170 degrees C for 2 hours, and it considered as the sample for an examination. 90-degree Peel peel strength and 180-degree Peel peel strength of this sample were measured at 23 degrees C.

<Solder thermal resistance> It laminated heating a polyimide film with an adhesives layer at 120 degrees C of rolling copper foil, heat hardening was carried out at 170 degrees C for 2 hours, and it considered as the sample for an examination. Temperature until it is immersed in the solder bath which fused this sample for 3 minutes and change (exfoliation, blistering, discoloration, etc.) arises in appearance was measured.

[0034] <Fire retardancy> It evaluated according to the fire-resistant test method of UL-94 specification.

<Heat-resistant adhesive property> 180-degree Peel peel strength when leaving the sample (for a copper foil adhesive property examination) produced by the adhesive examination under 120-degree-C atmosphere for 1000 hours was measured at 23 degrees C.

<Electric corrosion-proof nature> The adhesives constituent was applied so that hardening thickness might become the tandem type 80 micrometers, a line, and whose space are 120 micrometers to a copper substrate, and heat hardening was carried out at 170 more degrees C for 5 minutes by 140 degrees C for 2 hours, and it considered as the sample for an examination. The layer insulation resistance of the binder layer when leaving this sample under the atmosphere of 85% of humidity at 85 degrees C for 1000 hours and a copper substrate was measured.

[0035]

[Table 3]

表 3

試 料 No.		実施例 1	実施例 2	実施例 3	実施例 4	実施例 5	実施例 6	実施例 7	実施例 8	実施例 9
使用した重合体		A	B	C	D	E	F	G	H	I
配 合	重合体 (NV15%)	533	533	533	533	533	533	533	533	533
	フェノキシ樹脂 *9	-	-	-	-	-	-	-	-	-
	アクリロニトリル ブタジエンゴム *10	-	-	-	-	-	-	-	-	-
	ビスフェノールA型 エポキシ樹脂 *1	15	15	15	16	16	16	16	16	16
	クレゾールノボラック樹脂 *2	5	5	5	5	5	5	5	5	5
	エポキシ樹脂 *3									

(重量部)	フェノール樹脂 ^{*5}	8	8	8	8	8	8	8	8
	水酸化アルミ ^{*5}	85	85	85	85	85	85	85	85
	ビスフェニル型リン酸エステル ^{*6}	30	30	30	30	30	30	30	30
	メラミン樹脂 ^{*7}	10	10	10	10	10	10	10	10
	メラミンフェノール樹脂 ^{*8}	15	15	15	15	15	15	15	15
	イミダゾール系硬化促進剤 ^{*4}	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2

[0036]

[Table 4]

表 4

試料 No.		比較例1	比較例2	比較例3	比較例4	比較例5	比較例6	比較例7
使用した重合体		J	K	L	M	—	—	A
(重量部)	重合体(NV15%)	533	533	533	533	—	—	533
	フェノキシ樹脂 ^{*9}	—	—	—	—	80	—	—
	アクリロニトリル ブタジエンゴム ^{*10}	—	—	—	—	—	80	—
	ビスフェノールA型 エポキシ樹脂 ^{*1}	15	15	15	15	15	15	15
	クレゾールノボラック型 エポキシ樹脂 ^{*2}	5	5	5	5	5	5	5
	フェノール樹脂 ^{*3}	8	8	8	8	8	8	8
	水酸化アルミ ^{*5}	85	85	85	85	85	85	—
	ビスフェニル型リン酸エステル ^{*6}	30	30	30	30	30	30	—
	メラミン樹脂 ^{*7}	10	10	10	10	10	10	—
	メラミンフェノール樹脂 ^{*8}	15	15	15	15	15	15	—
	イミダゾール系硬化促進剤 ^{*4}	0.2	0.2	0.2	0.2	0.2	0.2	0.2

[0037]

[Table 5]

表 5

試料 No.		実施例1	実施例2	実施例3	実施例4	実施例5	実施例6	実施例7	実施例8	実施例9
接着性 (光沢面)	圧延鋼板 90°ピール強度	1.2	1.5	1.5	1.5	1.5	1.2	1.2	1.5	1.5
	180°ピール強度	1.2	1.5	1.6	1.5	1.5	1.3	1.2	1.5	1.5
	カプトン 90°ピール強度	0.9	1.1	1.1	1.1	1.1	0.9	1.1	1.0	1.1
	フィルム 180°ピール強度	1.0	1.1	1.1	1.1	1.1	1.0	1.1	1.0	1.1
はんだ耐熱性: °C		330	330	330	330	320	330	330	330	330
離層性		V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0
耐熱接着性: KN/n(180°ピール強度)		0.9	1.1	1.1	1.0	1.1	0.9	1.1	1.0	1.1
耐電食性: $\times 10^{11} \Omega$	初期	2.2	2.2	2.3	2.2	2.2	2.2	2.2	2.3	2.2
	(層間絶縁抵抗値) 電食試験1000h後	2.2	2.2	2.2	2.1	2.0	2.2	2.2	2.2	2.2

[0038]

[Table 6]

表 6

試 料 No.		比較例 1	比較例 2	比較例 3	比較例 4	比較例 5	比較例 6	比較例 7
接 着 性 : EN/h	圧延銅箔	90°ピール強度	0.8	1.2	0.8	1.3	1.0	1.1
	(光沢面)	180°ピール強度	1.0	1.2	0.9	1.3	1.1	1.2
	カプトン	90°ピール強度	0.7	1.0	0.3	0.9	0.3	1.0
	フィルム	180°ピール強度	0.8	1.0	0.4	1.0	0.5	1.0
は ん だ 耐 熱 性 : °C		270	260	330	300	260	260	320
難 燃 性		V-0	V-0	V-0	V-0	V-0	V-0	非燃焼
耐熱接着性 : EN/h(180°ピール強度)		0.5	0.2	0.4	0.5	0.2	0.2	1.0
耐電食性 : $\times 10^{-11} \Omega$ (層間絶縁抵抗値)	初 期	2.1	2.2	2.2	2.1	2.1	2.1	2.2
	電食試験1000h後	1.5	0.5	2.2	2.1	0.5	2.5	2.2

[0039]

[Effect of the Invention] Claims 1 and 2 and an adhesives constituent given in three are excellent in an adhesive property, thermal resistance, and electric corrosion-proof nature, and have fire retardancy by the non-halogen system.

[Translation done.]